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Memristive switching of single-component metallic nanowires

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Abstract
Memristors have recently generated significant interest due to their potential use in nanoscale logic and memory devices. Of the four passive circuit elements, the memristor (a two-terminal hysteretic switch) has so far proved hard to fabricate out of a single material. Here we employ electromigration to create a reversible passive electrical switch, a memristive device, from a single-component metallic nanowire. To achieve resistive switching in a single-component structure we introduce a new class of memristors, devices in which the state variable of resistance is the system’s physical geometry. By exploiting electromigration to reversibly alter the geometry, we repeatedly switch the resistance of single-component metallic nanowires between low and high states over many cycles. The reversible electromigration causes the nanowire to be cyclically narrowed to approximately 10 nm in width, resulting in a change in resistance by a factor of two. As a result, this work represents a potential route to the creation of nanoscale circuits from a single metallic element.

1. Introduction
Metals have long been used to form resistors, capacitors, and inductors—the three classical passive circuit elements in which geometry determines the principle behavior of the device [1]. A fourth passive circuit element, the memristor [2, 3], has generated significant recent interest [4] due to its potential use in nanoscale logic and memory devices. Among the presently demonstrated memristors are those whose resistance depends on state variables such as coupled ionic and electronic conduction [4–6], phase transitions [7, 8], redox reactions in organic semiconductors [9, 10], and the configuration of molecular heterostructures [11–14]. As with most solid-state electrical switches, the memristors reported so far comprise multiple components and interfaces. Here we introduce a new class of memristors and electrical switches: devices that can be constructed from a single component and in which the state variable of resistance is the system’s physical geometry. Using electromigration, we reversibly switch the resistance of single-component metallic nanowires between low and high states over many cycles. This work thus completes the array of fundamental passive circuit elements, now including the memristor, which can be fabricated from a single metallic material.

Since the first developments of integrated circuits (ICs), electromigration has been known to degrade the structures and interconnects of their components, thereby limiting their useful lifetime [15]. In recent years however, electromigration, once the bane of the microelectronics community, has become a topic of intense interest due to its potential in fabricating nanoscale structures at size scales much smaller than is possible with nanolithography techniques [14, 16–18]. Recent advances utilizing feedback controlled electromigration (FCE) have yielded reproducible methods for forming ∼1 nm scale features by utilizing feedback control to prevent the thermal runaway typically associated with electromigrating nanoscale wires [19–24]. Most of this work has investigated electromigration on the nanoscale with the objective of utilizing it for the fabrication of electrical devices at ever-smaller sizes. Here, we utilize electromigration itself as the nanoscale electrical switch by demonstrating significant reversible switching of the resistance (the memristive effect) in single-component metallic nanowires.

2. Experimental details
All samples were fabricated using a Raith eLine electron-beam lithography (EBL) system. A bi-layer resist consisting of
methyl-methacrylate/methacrylic-acid (MMA/MAA) copolymer and poly(methyl-methacrylate) (PMMA) were spun onto oxidized silicon substrates (300 nm thermal oxide). Following EBL exposure, samples were developed in a mixture of 3:1 isopropanol:methyl-2-pyrrolidinone (NMP) held at 70°C for approximately 1 h.

Electrical measurements were carried out at room temperature using a custom-built micro-probe station and voltage-clamp circuit. Current was measured using a Keithley 6517A electrometer and all data were acquired using LabVIEW.

The devices consist of 20 nm thick gold nanowires with an initial width of ~100 nm and length of ~300 nm. The lower inset in figure 1(a) shows a scanning-electron microscope (SEM) image of a typical nanowire structure. Each nanowire is connected to two large leads that allow the current to flow. Two additional smaller leads stem off of the current leads and allow for the application of an applied bias voltage to the nanowire. All metallic nanowires were constructed without Ti or Cr adhesion layers, as is commonly employed in electromigrated nanogap work [17, 25], thus making the nanowires truly single component.

Due to the relatively high electrical conductance of the nanowire compared to the leads, electrical measurements were carried out using a voltage-clamp circuit [26] (similar to a four-terminal circuit utilized recently for FCE [22]), illustrated schematically in figure 1(a) (upper inset). The operational amplifier (op-amp) maintains the applied voltage \( V \) across the nanowire, which has a variable resistance \( R_N \), by sourcing current from its output with a variable voltage \( V_{tot} \). Since negligible voltage is dropped across the lead resistances tied into the op-amp inputs, \( V \) appears across the metallic nanowire regardless of its resistance, \( R_N \). Essentially, the voltage-clamp circuit takes the place of slower feedback algorithms used in the past to control electromigration [21, 22]. The voltage clamp controls the electromigration by forcing the power dissipation to decrease as the nanowire resistance increases during electromigration (for a fixed applied voltage), which works to avoid thermal runaway.

3. Results and discussion

In figure 1(a) we show the conductance of the nanowire as a function of applied voltage, which is ramped with a single linear sweep over a period of 600 s. When the applied voltage is ~150 mV, the nanowire conductance drops significantly due to electromigration. Figure 1(b) shows the total conductance of the complete nanowire circuit as a function of total voltage \( V_{tot} \) applied to the circuit, including the lead resistances \( R_L \). The data exhibit the typical ‘C’-shaped curves commonly observed when taken using software algorithms of FCE [21], illustrating that the voltage-clamp circuit works in the same manner. Moreover, the critical current density obtained in the nanowire at the onset of electromigration is roughly \( 5 \times 10^{12} \) A m\(^{-2}\), in close agreement with previous work [27].

In order to observe memristance in our nanowire devices, we exploit the symmetric nature of the electromigration process [16]. During an initial voltage ramp, a critical power dissipation is reached in the wire and electromigration begins which causes the conductance to decrease. The voltage is then reduced and ramped in the opposite direction. At roughly the same critical power dissipation corresponding to a negative applied voltage, electromigration begins again but now acts to increase the wire conductance as voids created during the initial voltage ramp are refilled. Figure 2(a) shows the current–voltage \( (I–V) \) curve of this device as a function of the applied \( V \) for one complete cycle in which a void is formed and then refilled. Figure 2(b) illustrates this effect schematically.

A key to observing and controlling the memristance of our electromigration-based device is the implementation of the voltage-clamp circuit (figure 1(b)). The voltage clamp allows for the monitoring and control of the memristance of the nanowire without the effect being obscured by the typical lead resistances (~200 \( \Omega \)) in the system. In the inset of figure 2(a) we show data corresponding to the same memristance cycle in figure 2(a), but instead plot the current as a function of \( V_{tot} \), the voltage that would need to be applied to the entire circuit if the voltage clamp were not used. Clearly, the memristance effect is strongly obscured by the lead resistance.

Figure 1. (a) Nanowire conductance as a function of the applied voltage. The insets show a schematic diagram of the feedback circuit and a SEM image of a typical device structure. (b) Total current and conductance of the nanowire circuit as a function of total voltage applied to the circuit including the lead resistances, \( R_L \), showing the ‘C’ shaped curves typical of feedback controlled electromigration [21].
Figure 2. (a) $I-V$ characteristics of a nanowire over one voltage ramp cycle showing a significant hysteresis, or memristance. The inset shows the same data plotted against $V_{tot}$, the voltage applied across the entire circuit, which shows little hysteric effect. (b) Schematic illustration of an Au nanowire before (upper) and after (lower) electromigration illustrating how geometrical changes lead to resistance changes.

This memristive effect in metallic nanowires can be reproduced over many cycles. Figure 3(a) shows the $I-V$ curves of 14 consecutive cycles of a nanowire having an applied sawtooth voltage as a function of time, with the corresponding conductance and applied voltage shown in figure 3(b). The $I-V$ curves show a clear reproducible memristance effect [4] with an on/off ratio of $\sim 2$, as shown in figure 3(b). We find that such repeatability is typical in our devices, and that devices may be disconnected and reconnected without altering the devices’ memristive properties. We also generally find that the reproducibility of this memristive effect improves after the first few cycles of the electromigration of a nanowire. This improvement in reproducibility is likely due to the completion of restructuring of grain boundaries within the nanowire region that occurs during current-induced Joule heating in the early stages of resistive switching. Although figure 3 only shows switching over 14 complete cycles, our current nanowires can typically endure dozens of more cycles before failure. After repeated cycling the on/off ratios of our current nanowires can shift and become unstable as the nanowire geometry is reversibly cycled.

The observed memristive effect is likely due to the induced temperature gradients along the nanoscale wire. The metallic atom flux per unit area due to electromigration in the nanowire can be described by the one-dimensional mass transport relation, $U = ND(T)Z^e j \rho / k_B T$ [15], where $N$ is the atom concentration, $D(T)$ is the temperature ($T$) dependent diffusion coefficient, $Z^e$ is the effective charge of the metallic atoms, $j$ is the applied electrical current density, $\rho$ is the electrical resistivity, and $k_B$ is Boltzmann’s constant. A void is formed at locations where the divergence, $\frac{\partial U}{\partial T} \frac{\partial x}{\partial t} > 0$, is greater than zero while an accumulation of mass occurs at locations where the divergence is less than zero. At the two ends of the nanowire, adjacent to the large leads, the temperature gradients have opposite signs. Since $\frac{\partial U}{\partial T}$ is a monotonically increasing function of $T$ over a realistic temperature range (for $Z^e j > 0$), the divergence should have opposite signs at the two ends of the nanowires, which thus drives the void formation and mass accumulation represented in figure 2(b). Since $\frac{\partial U}{\partial T}$ changes sign upon reversal of $j$, a void can be made to refill by reversing the direction of the applied electrical current. This cyclical void formation and refilling is the likely root of the memristive effect observed in our nanowire devices.

The minimum nanowire width during a memristive cycle can be estimated from the conductance value at its low state. Assuming a quantum of conductance ($G_0 = 7.75 \times 10^{-5}$ S) for each atomic channel and a diameter for gold atoms of $\sim 0.3$ nm yields a cross sectional area of approximately $35$ nm$^2$. For the
wire at its narrowest point. If the thickness of the wire is approximately 3 nm, as suggested by recent electromigration work [16], then this would correspond to a wire width of about 12 nm. To achieve the on/off conductance values in figure 3(b) would require the movement of at least 40 000 Au atoms, assuming that the length of the electromigrated region of the wire is approximately 20 nm long [16]. Considerably lower conductance values, and as a result greater on/off ratios, could be achieved if this minimum wire dimension is decreased further. Moreover, a narrower wire would require the movement of much less material and could thus significantly increase switching times.

The ultimate potential switching speed for an electromigration-based memristor can be extrapolated from the behavior of our current devices. The timescale for switching from a high to a low state in figure 3(b) is roughly 10 s. If this switching time scales linearly with the number of atoms moved (~40 000) and the length of the nanowire (~200 nm), we could expect a roughly 10 μs response time if the electromigrated void is operated on the few atom size and the overall length of the nanowire is reduced to 10 nm in length. A further significant reduction in switching time could be achieved by operating at higher biases, since this would increase the temperature in the nanowire and, as a result, significantly increase the mobility of the atoms. Since the mobility of atoms generally have a strong exponential dependence on the temperature [15], the timescale for electromigration could be decreased by many orders of magnitude by operating at higher biases. In such a scenario, the likely limit to operating speed will be the thermal time constant which dictates the timescale over which the temperature and temperature gradients within the nanowire are increased. Recent estimates for ~100 nm sized metallic wires suggest that this time constant is less than a nanosecond [28], with smaller sized nanowires likely to have an even smaller time constant.

4. Conclusions

In summary, we have developed a new type of memristor—one whose resistance changes with geometry through electromigration. The devices can be reversibly switched between low and high conductance values corresponding to the cyclical narrowing of the nanowire to approximately 10 nm in width. Since the effect is due to geometrical changes, the devices can be constructed from a single component. As a result, the devices present a potential route to the creation of nanoscale circuits comprising a single metallic element. Due to the prevalence of electromigration in conducting materials and the fact that the memristance depends on its geometrical form, we also expect that this effect could be useful in a wide assortment of nanoscale conductors.

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